

REMARKS

The applicant respectfully requests reconsideration in view of the amendments and the following remarks. The applicant has amended claim 1 in order to overcome the 35 U.S.C. 112, second paragraph rejection.

Claims 1-6, 8-17, 19, 20 and 22 are rejected under 35 U.S.C. 112, second paragraph as being indefinite for failing to particularly point out and distinctly claim the subject matter which applicant regards as the invention. Claims 1-6, 8-17, 19, 20 and 22 are rejected under 35 U.S.C. 103(a) as being unpatentable over US Patent 6,562,971 ("Frauenkron") in view of US Patent 6,350,874 ("Ogawa"). The applicant respectfully traverses these rejections.

The applicant appreciates that the Examiner has withdrawn the double patenting rejection and the 35 U.S.C. 102 rejections.

35 U.S.C. 112, Second Paragraph Rejection

Claims 1-6, 8-17, 19, 20 and 22 are rejected under 35 U.S.C. 112, second paragraph as being indefinite for failing to particularly point out and distinctly claim the subject matter which applicant regards as the invention. The applicant appreciates the Examiner pointing out this error in claim 1. The applicant has amended claim 1 as suggested by the Examiner. For the above reasons, this rejection should be withdrawn.

35 U.S.C. 103 Rejection

Claims 1-6, 8-17, 19, 20 and 22 are rejected under 35 U.S.C. 103(a) as being unpatentable over Frauenkron in view of Ogawa. The present invention relates to a method of

increasing the cutting hardness of a shaped body (see the specification at page 1, lines 1-8, and claim 1).

The applicant requires the following mandatory step for the preparation of the shaped body:

- a) a crystalline aluminosilicate having a SiO₂/Al₂O₃ molar ratio in the range of 10:1 to 1200:1
- b) a hinder selected from among oxides of silicon and/or zirconium
- c) calcining at temperatures from 100 to 600°C and
- d) treating the calcined shaped body with a gas consisting of water vapor at temperatures from 100 to 600°C and an absolute pressure of from 0.1 to 10 bar for a period of at least 20 hours (see claim 1).

It is an object of the applicant's invention to discover an improved method of increasing the cutting hardness of a shaped body comprising a crystalline aluminosilicate for use as catalyst in known zeolite-catalyzed reactions, in particular for use as catalyst in a process for preparing triethylenediamine (TEDA) by reaction of ethylenediamine (EDA) and/or piperazine (PIP), which is simple to carry out and results in an increased catalyst operating life and catalyst stability compared to the prior art. At the same time, no deactivation of catalyst should occur as a result of the method and the selectivity in the relevant zeolite-catalyzed reactions, in particular the selectivity to TEDA in the abovementioned process for preparing TEDA, should not be adversely affected (see page 2, lines 23-28 of the present invention).

The cutting hardness of a shaped body is a measure of its mechanical stability. For industrial applications it is generally desirable for the shaped catalyst bodies to have a cutting hardness of at least 9.8 N (see page 2, lines 33-37 of the present invention).

The above described object is achieved by the method according to claim 1. The present

invention overcomes the aforementioned problems of catalyst destruction by increasing the cutting hardness of the shaped catalyst bodies prior to the installation into the reactor. By the increased cutting hardness of the catalyst according to the present invention, the pulverization and fragmentation during the installation process of the catalyst into the reactor is minimized. This effect is achieved, by calcining as shaped body, comprising a crystalline alumo silicate, obtaining a $\text{SiO}_2/\text{Al}_2\text{O}_3$ molar rating in the range of 10:1 to 1200:1, according to step d) of the present invention, the treating of the calcined catalyst body with a gas consisting of water vapor at temperatures from 100 to 600 °C and an absolute pressure of from 0.1 to 10 bar for a period of at least 20 hours.

Frauenkron discloses a method of forming a body which involves the following steps (see column 10, line 61; column 11, line 64 to column 12, line 2; example in column 14, line 65 to column 15, line 6):

- a) mixing a zeolite powder with silica as binder, having a $\text{SiO}_2/\text{M}_2\text{O}_3$ ratio of greater than 1400:1 to 40000:1 (see claim 1)
- b) extruding the mixture
- c) calcining the formed body at 250 to 950°C for 2 to 5 hours (in example 1: 500°C for 5 hours),

According to Frauenkron the shaped body is subsequently used as catalyst for the synthesis of TEDA out of a feed comprising EDA:PIP:H₂O (35:15:50) (see example 1).

The important step d) of the present invention - the treatment with a gas consisting of water vapor at 100 to 600°C and an absolute pressure of from 0.1 to 10 bar for a period of at least 20 hours - after calcining the shaped body is not disclosed by Frauenkron. Therefore, Frauenkron

does not teach a method of increasing the cutting hardness of a shaped catalyst body prior to the use of the catalyst in a reactor. Frauenkron gives no hint of how to increase the cutting hardness of a catalyst body. This is substantiated by the fact that Frauenkron does not provide data of the cutting hardness of the catalyst described. Furthermore, the word “cutting hardness” does not appear in the description of Frauenkron. As a result, the applicant believes that a shaped catalyst body produced by the method described by Frauenkron will be destroyed by the mechanical forces during the installation process of the catalyst into the reactor. Consequently, the disclosure of Frauenkron does not overcome the problems of the prior art.

The argument of the Examiner in the first paragraph of page 4 of the Office Action, that the shaped body according to Frauenkron is treated by exposure to a gas including water vapor PIP and EDA at temperatures of 345 °C (column 15, lines 10-20) seems to be of little relevance.

First of all, Frauenkron gives no proof that an increase of cutting hardness of a catalyst body takes place by this treatment. Secondly, even if the increase of cutting hardness would occur, no hint for this increasement is given in the disclosure of Frauenkron. Thirdly, the destruction of the catalyst during the installation of the catalyst into the reactor would irrecoverably have taken place. Therefore, the teaching of Frauenkron cannot overcome the problems of the prior art.

Frauenkron differs from the present invention by the features of 1) step d) which causes an increasing of the cutting hardness of the catalyst body and 2) the SiO₅/Al₂O₃ molar ratio of the crystalline aluminosilicate.

In the last response, the applicant pointed out that Frauenkron taught away from the claimed molar ratio.

Frauenkron discloses at col. 8, lines 10-39:

The zeolite employed as catalyst in the process according to the invention for the preparation of TEDA has a skeletal structure which principally consists of silicon dioxide (SiO_2).

Besides silicon dioxide, the zeolite may also contain traces of other divalent, trivalent or tetravalent metals **M (oxidation state of the metals II, III or IV)** in the form of metal oxides $\text{M}_{2/z} \text{O}$ ($z=2, 3$ or 4), where the molar ratio of silicon dioxide and the metal oxide, $\text{SiO}_2 / \text{M}_{2/z} \text{O}$ (this molar ratio is also known as the 'modulus') is greater than 10 for tetravalent metals ($z=4$), greater than 100 for divalent and trivalent metals ($z=2$ or 3) and greater than 1400 for $\text{M}=\text{Al}(z=3)$.

The zeolite catalysts preferably used in the process according to the invention have, for $\text{M}=\text{Al}$, an $\text{SiO}_2 / \text{M}_2 \text{O}_3$ molar ratio of from greater than 1400 to 40,000:1, in particular from greater than 1400 to 5000:1, for $\text{M}=\text{metal}$ in oxidation state II or $\text{M}=\text{two or more metals}$ in oxidation state II, have an SiO_2 / MO molar ratio of from greater than 100 to 40,000:1, in particular from greater than 200 to 5000:1, for $\text{M}=\text{metal}$ in oxidation state III or $\text{M}=\text{two or more metals}$ in oxidation state III, have an $\text{SiO}_2 / \text{M}_2 \text{O}_3$ molar ratio of from greater than 100 to 40,000:1, in particular from greater than 200 to 5000:1, and for $\text{M}=\text{metal}$ in oxidation state IV or $\text{M}=\text{two or more metals}$ in oxidation state IV, have an $\text{SiO}_2 / \text{MO}_2$ molar ratio of from greater than 10 to 40,000:1, in particular from greater than 25 to 5000:1. (emphasis added)

For Al_2O_3 , Frauenkron teaches that the molar ratio of $\text{SiO}_2 / \text{Al}_2\text{O}_3$ is from greater than 1400 to 40,000:1, in particular from greater than 1400 to 5000:1. Frauenkron's minimum ratio is **greater than 1400** while the applicant's claimed maximum ratio is 1200. Therefore, Frauenkron teaches away from the applicant's claimed invention ($\text{SiO}_2 / \text{Al}_2\text{O}_3$ molar ratio of 10:1 to 1200:1) and teaches to have at least a molar ratio of $\text{SiO}_2 / \text{Al}_2\text{O}_3$ being greater than 1400.

The Examiner now has relied upon Ogawa for the teaching of the claimed ratio. Ogawa discloses a method for producing triethylene diamine or piperazine by the use of a catalyst with a molar ratio of $\text{SiO}_2 / \text{Al}_2\text{O}_3$ of 40 to 5,000 (see abstract and col. 4, lines 15-18). However, Ogawa

gives no hint to treat a calcined shaped body comprising a crystalline aluminosilicate having a $\text{SiO}_2/\text{Al}_2\text{O}_3$ molar ratio in the range of 10:1 to 1200:1 with a gas consisting of water vapor by temperatures from 100 to 600°C. Ogawa therefore also gives no hint to a method for increasing the cutting hardness of a shaped body.

Furthermore, contrary to what the Examiner has alleged, there would be reason for a person of ordinary skill in the art to use a molar ratio of $\text{SiO}_2/\text{Al}_2\text{O}_3$ less 1400 when Frauenkron specifically states that the minimum ratio is 1400. For the above reasons, this rejection should be withdrawn.

In view of the above amendment, applicant believes the pending application is in condition for allowance.

Applicant believes no fee is due with this response. However, if a fee is due, please charge our Deposit Account No. 03-2775, under Order No. 13156-00177-US from which the undersigned is authorized to draw.

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Respectfully submitted,

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